

Dynamical Structure Factors for the Heisenberg Chain with Alternating Exchange

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Abstract

We discuss the transition strength between the disordered ground state and the basic low-lying triplet excitation for the Heisenberg chain with alternating exchange based on series expansions in the interdimer interaction up to 10th order. We demonstrate that the wave vector dependence of the simple dimer approximation is strongly modified in higher orders.

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Introduction

Low-dimensional quantum antiferromagnets have received much interest in recent years since they serve as model substances allowing to investigate in detail the effects of quantum fluctuations and to test theoretical models.

One important class of materials in this context consists of an assembly of dimers (two strongly coupled spins $1/2$) which interact sufficiently weak to guarantee that the dimer gap does not close. These materials are characterized by a disordered singlet ground state and a finite spin gap to triplet excited states. A number of investigations has been performed on the simplest model systems of this type, the Heisenberg chain with alternating exchange: The dispersion of the lowest singlet-triplet transition as well as the two particle bound states have been calculated by series expansions in the interdimer interaction up to 10th order^{1,2,3,4} and the corresponding structure factor $S(q)$ up to third order². Here we supplement the analysis of Ref. 1 by presenting cluster expansion results for the dynamic structure factor up to 10th order.

Since the lowest triplet mode is an isolated excitation, the weight of the corresponding δ -function, i.e. the contribution of the singlet-triplet transition to the structure factor $S(\mathbf{q})$ is the quantity to be discussed. We will use the following notations: The dynamical structure factor for spins localized on a Bravais lattice is defined as

$$S^{\alpha\beta}(\mathbf{q}, \omega) = \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle \mathcal{S}^{\alpha}(\mathbf{q}) \mathcal{S}^{\beta}(-\mathbf{q}) \rangle \quad (1)$$

where

$$\mathcal{S}^{\alpha}(\mathbf{q}) = \sum_{\mathbf{R}} e^{-i\mathbf{q}\mathbf{R}} \mathcal{S}^{\alpha}(\mathbf{R}) \quad (2)$$

is the Fourier transformation of the spin operators at lattice sites \mathbf{R} . The superscripts α, β denote the spin components and the brackets $\langle \dots \rangle$ thermal expectation values (which for $T=0$ reduce to groundstate expectation values $\langle 0 | \dots | 0 \rangle$).

If we consider transitions from the groundstate to some isolated eigenstate $|n\rangle$ of \mathcal{H} with energy $\omega_n(\mathbf{q})$, we obtain δ -peaked contributions to the dynamical structure factor

$$S^{\alpha\beta}(\mathbf{q}, \omega) = \sum_n \langle 0 | \mathcal{S}^{\alpha}(\mathbf{q}) | n \rangle \langle n | \mathcal{S}^{\beta}(-\mathbf{q}) | 0 \rangle \delta(\omega - \omega_n(\mathbf{q})) \quad (3)$$

$$= \sum_n I_n^{\alpha\beta}(\mathbf{q}) \delta(\omega - \omega_n(\mathbf{q})). \quad (4)$$

Inelastic neutron scattering probes directly the transition from the (singlet) groundstate to the lowest (triplet) excitation $|t\rangle$ of the quantum disordered compound and we will reduce our discussion to this contribution to the dynamical structure factor. Owing to the rotational symmetry of the model it is sufficient to calculate $I_t^{zz}(\mathbf{q})$ only and we use the shorthand $I(\mathbf{q}) := I_t^{zz}(\mathbf{q}, 1)$.

In the following we will give theoretical results for the 1D array of interacting dimers. This model was treated before², it is, however, instructive (i) to demonstrate for the 1D case, that the simple lowest order results are modified by further terms which emerge in higher orders and (ii) to present the quantitative changes for the transition strength comparing first order to 10th order results. The hamiltonian of this model is of the following form

$$\mathcal{H} = J \sum_{i=0}^{N/2} \mathbf{S}_{2i} \mathbf{S}_{2i+1} + \lambda \mathbf{S}_{2i+1} \mathbf{S}_{2i+2}, \quad J > 0. \quad (5)$$

Here, N denotes the number of spins and periodic boundary conditions are used. In the case of $\lambda = 0$ the groundstate of the system consists of rung singlets on the bonds $(2i, 2i+1)$. Local rung singlets can be excited to rung triplets which become gapped dispersive excitations when switching on λ , $0 < \lambda < 1$. In the limit $\lambda = 1$ we arrive at the well known Heisenberg antiferromagnet (HAFM) where the lowest, gapless excitations are pairs of $S = 1/2$ spinons.

The one magnon excitation energies $\omega(\mathbf{q})$ have been obtained by perturbation expansion in λ up to 9th order in Ref. 2 and to 10th order in Ref. 1 using the cluster expansion approach.

The dynamical structure factor

For the chain geometry shown in Fig. 1 we get for the intensity up to first order in λ the following expression

$$I(\mathbf{q}) = \sin^2 \frac{\mathbf{q}\mathbf{d}}{2} \left(1 + \frac{1}{2} \lambda \cos(qa) \right) + \mathcal{O}(\lambda^2). \quad (6)$$

Here, q is the projection of the wavevector \mathbf{q} in the chain direction. We note that the term $\propto \sin^2(\mathbf{q}\mathbf{d}/2)$ is typical for systems consisting of isolated dimers and is known as the dimer structure factor. The first order correction in λ adds an additional modulation to the intensity, which depends on the ratio $\sigma = \|\mathbf{d}\|/\|\mathbf{a}\|$.

Using the cluster expansion method (see Appendix A) we have systematically calculated the series in λ for the intensity up to 10th order. This requires linked clusters consisting of

maximum 10 bonds. The resulting series can be split into three different terms:

$$I(\mathbf{q}) = B_c(\mathbf{q}, \lambda) + B_s(\mathbf{q}, \lambda) + \Lambda(\mathbf{q}, \lambda). \quad (7)$$

To illustrate the result we give the series up to fourth order¹⁰:

$$\begin{aligned} B_c(\mathbf{q}, \lambda) &= \sin^2 \frac{\mathbf{q}\mathbf{d}}{2} \sum_{j=0}^4 \mu_j \cos(jqa) \\ B_s(\mathbf{q}, \lambda) &= \sin \mathbf{q}\mathbf{d} \sum_{j=0}^4 \nu_j \sin(jqa) \end{aligned} \quad (8)$$

where

$$\mu_0 = 1 - \frac{5}{16}\lambda^2 - \frac{3}{32}\lambda^3 + \frac{25}{1536}\lambda^4, \quad \mu_1 = \frac{1}{2}\lambda - \frac{1}{8}\lambda^2 - \frac{5}{192}\lambda^3 + \frac{41}{2304}\lambda^4, \quad (9)$$

$$\mu_2 = \frac{3}{16}\lambda^2 + \frac{7}{48}\lambda^3 + \frac{23}{1024}\lambda^4, \quad \mu_3 = \frac{5}{64}\lambda^3 + \frac{155}{2304}\lambda^4, \quad \mu_4 = \frac{35}{1024}\lambda^4, \quad (10)$$

$$\nu_0 = 0, \quad \nu_1 = \frac{1}{8}\lambda^2 + \frac{7}{192}\lambda^3 - \frac{131}{4608}\lambda^4, \quad \nu_2 = \frac{1}{96}\lambda^3 + \frac{25}{4608}\lambda^4, \quad (11)$$

$$\nu_3 = \frac{23}{2304}\lambda^4, \quad \nu_4 = 0, \quad (12)$$

and

$$\Lambda(\mathbf{q}, \lambda) = \frac{1}{128}\lambda^4 (\cos(\mathbf{q}\mathbf{d}) - \cos(2qa)). \quad (13)$$

The terms $B_c(\mathbf{q}, \lambda)$ are consistent with previous publications², whereas $B_s(\mathbf{q}, \lambda)$ and $\Lambda(\mathbf{q}, \lambda)$ are additional corrections. They originate from a complete expansion of both the ground state $|0\rangle$ and the first excited state $|t\rangle$. If one assumes that $\langle 0|S_{2i}|t\rangle = -\langle 0|S_{2i+1}|t\rangle$ for the matrix elements on even and odd sites, one ends up with $B_c(\mathbf{q}, \lambda)$ only. However this is only correct to first order and in general we have $\langle 0|S_{2i}|t\rangle \neq -\langle 0|S_{2i+1}|t\rangle$. This is related to virtual states which occur during the perturbation expansion and exhibit odd parity under the exchange of the triplets. These corrections begin to appear in second order.

In Figs. 2 and 3 we show some typical plots of the intensity $I(\mathbf{q})$ for two different ratios $\sigma = 10/15$, $10/20$ and two different coupling strength $\lambda = 0.4$, 0.8 . The difference between the zeroth and the first order emerges very clearly. The higher order terms emphasize the modulation originating from the two length scales $\|\mathbf{a}\|$ and $\|\mathbf{d}\|$.

Sum rule

Independently of whether the magnetic system is ordered or not, the total integrated scattering intensity has a well defined magnitude, determined by the local spin length through the following sum rule:

$$\mathcal{I} = \sum_{\alpha} \frac{\int d\mathbf{q} \int \frac{d\omega}{2\pi} S^{\alpha\alpha}(\mathbf{q}, \omega)}{\int d\mathbf{q}} = S(S+1). \quad (14)$$

In the case of the one dimensional alternating chain the integral reduces to the constant part μ_0 , since only non oscillating terms will survive the integration:

$$\mathcal{I} = \frac{3}{4} \left(1 - \frac{5}{16}\lambda^2 - \frac{3}{32}\lambda^3 + \frac{25}{1536}\lambda^4 + \dots \right) \leq \frac{3}{4} \quad S = \frac{1}{2}, \quad \lambda \ll 1. \quad (15)$$

For the noninteracting case ($\lambda = 0$) the sum rule is exhausted by the one triplet excitation since it is an exact eigenstate: From (3) we see that this excitation gives the only non-vanishing matrix element. Switching on the coupling between the dimers, more and more intensity goes in two or more magnon scattering processes.

Conclusion

Using the $S = \frac{1}{2}$ Heisenberg chain with alternating exchange interactions we have demonstrated by high order series expansions that the simple dimer structure factor is considerably modified in higher orders in the interdimer exchange. In a forthcoming publication we will extend our approach to cover interacting dimers in a three-dimensional lattice in order to allow a discussion of the transition strength for the interacting dimer materials KCuCl_3 and TlCuCl_3 . These materials have been investigated in detail in the last years in inelastic neutron scattering (INS) experiments^{5,6}, thus directly exploring the basic singlet-triplet transition in all of reciprocal space.

APPENDIX A: CLUSTER EXPANSION

In this appendix we briefly summarize the method of cluster expansion for the dynamical structure factor in the case of the alternating chain. Some detailed considerations can be

found elsewhere⁷. As shown in FIG. 1 the cristallographic unit cell contains two spins. Thus the Fourier transform of the spin operator splits into two parts and reads as:

$$S^\alpha(\mathbf{q}) = \sum_n e^{-iqna} (e^{-i\mathbf{q}\mathbf{d}} S_1^\alpha(n) + e^{i\mathbf{q}\mathbf{d}} S_2^\alpha(n)). \quad (\text{A1})$$

As before $\|\mathbf{a}\| = a$ is the distance between neighbouring spins and $2\mathbf{d}$ denotes the spacing between the two spins on a dimer. In our notation q is the projection of the wave vector \mathbf{q} on the chain direction.

Using translational invariance with respect to the center of the dimer we obtain for the singlet-triplet transition amplitude:

$$I(\mathbf{q}) = \sum_n e^{-iqan} \left[A_{11}^{zz}(n) + A_{22}^{zz}(n) + e^{2i\mathbf{q}\mathbf{d}} A_{12}^{zz}(n) + e^{-2i\mathbf{q}\mathbf{d}} A_{21}^{\alpha\beta}(n) \right] \quad (\text{A2})$$

where

$$A_{ij}^{zz}(n) = \langle 0 | \mathcal{S}_i^z(0) | 1 \rangle \langle 1 | \mathcal{S}_j^{z\dagger}(n) | 0 \rangle, \quad i, j = 1, 2. \quad (\text{A3})$$

Here, the sum is taken over all integer numbers. However, it is more convenient to calculate the functions $A_{ij}^{zz}(n)$ for positive numbers n . This is feasible by the benefit of inversion symmetry concerning a dimer center of mass which is manifest in the relations:

$$A_{11}^{zz}(-n) = A_{22}^{zz}(n) \quad \text{and} \quad A_{12}^{zz}(-n) = A_{21}^{zz}(n). \quad (\text{A4})$$

Inserting (A4) into (A2) one arrives at the following result:

$$\begin{aligned} I(\mathbf{q}) = & 2 \sum_{n>0} (A_{11}^{zz}(n) + A_{22}^{zz}(n)) \cos(qna) \\ & + 2 \sum_{n>0} (A_{12}^{zz}(n) \cos(qna - 2\mathbf{q}\mathbf{d}) + A_{21}^{zz}(n) \cos(qna + 2\mathbf{q}\mathbf{d})) \\ & + A_{11}^{zz}(0) + A_{22}^{zz}(0) + e^{2i\mathbf{q}\mathbf{d}} A_{12}^{zz}(0) + e^{-2i\mathbf{q}\mathbf{d}} A_{21}^{zz}(0). \end{aligned} \quad (\text{A5})$$

We state that the functions $A_{ij}^{zz}(n)$ have to be calculated for positive n only.

At first glance the functions $A_{ij}^{zz}(n)$ are groundstate expectation values which can be computed by well known cluster expansion methode⁸. The projection operator $\mathcal{P} = |1\rangle\langle 1|$ has to be evaluated from the one magnon states $|\psi^{(i)}\rangle$ where i labels the lattice site. By means of degenerate cluster expansion these states are generated order by order⁹. Then we find for projection operator:

$$\mathcal{P} = |1\rangle\langle 1| = \sum_{ij} (g^{-1})_{ij} |\psi^{(i)}\rangle \langle \psi^{(j)}|. \quad (\text{A6})$$

g is the overlapping matrix of the $|\psi^{(i)}\rangle$:

$$g_{ij} = \langle \psi^{(i)} | \psi^{(j)} \rangle. \quad (\text{A7})$$

To invert g we use the fact that g is the unit matrix for $\lambda \rightarrow 0$:

$$g = I + \tilde{g}. \quad (\text{A8})$$

Owing to the matrix norm $\|\tilde{g}\| < 1$ we apply a geometric series to invert g :

$$(I + \tilde{g})^{-1} = \sum_{i=0}^{\infty} \tilde{g}^i. \quad (\text{A9})$$

Now we have everything at hand to calculate the singlet-triplet-intensity of the dynamical structure factor: Apply degenerate perturbation theory to obtain the states $|\psi^{(j)}\rangle$ and \mathcal{P} . Then calculate g and invert this matrix by using (A9). Finally, apply non degenerate perturbation theory to compute the functions $A_{ij}^{zz}(n)$.

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¹⁰ Higher order terms are available on request; more details can be found in M. Müller, PhD-Thesis, Universität Hannover (2002).

FIGURES

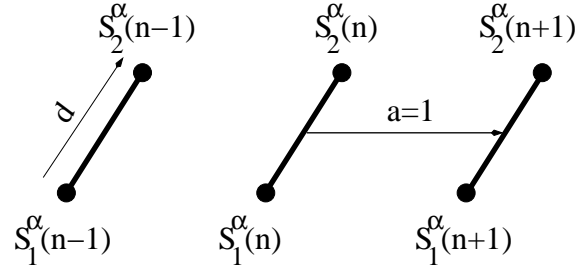


FIG. 1: Arrangement of dimers and labeling of spins of the spin ladder

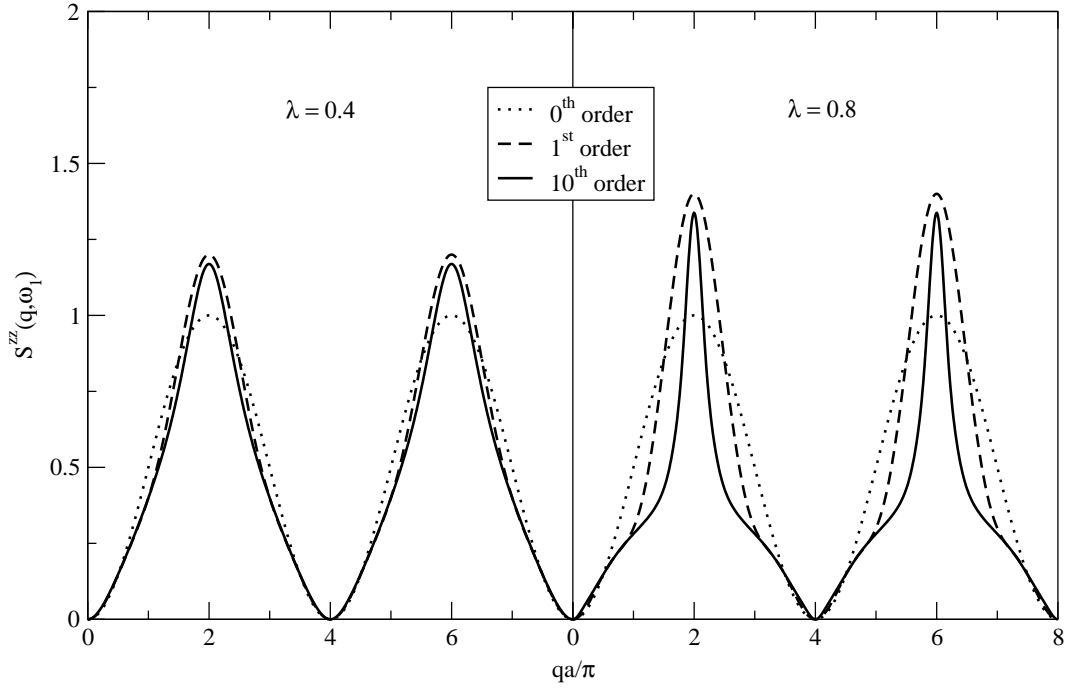


FIG. 2: Structure factor with $\lambda = 0.4, 0.8$ and ratio $d/a = 10/20$

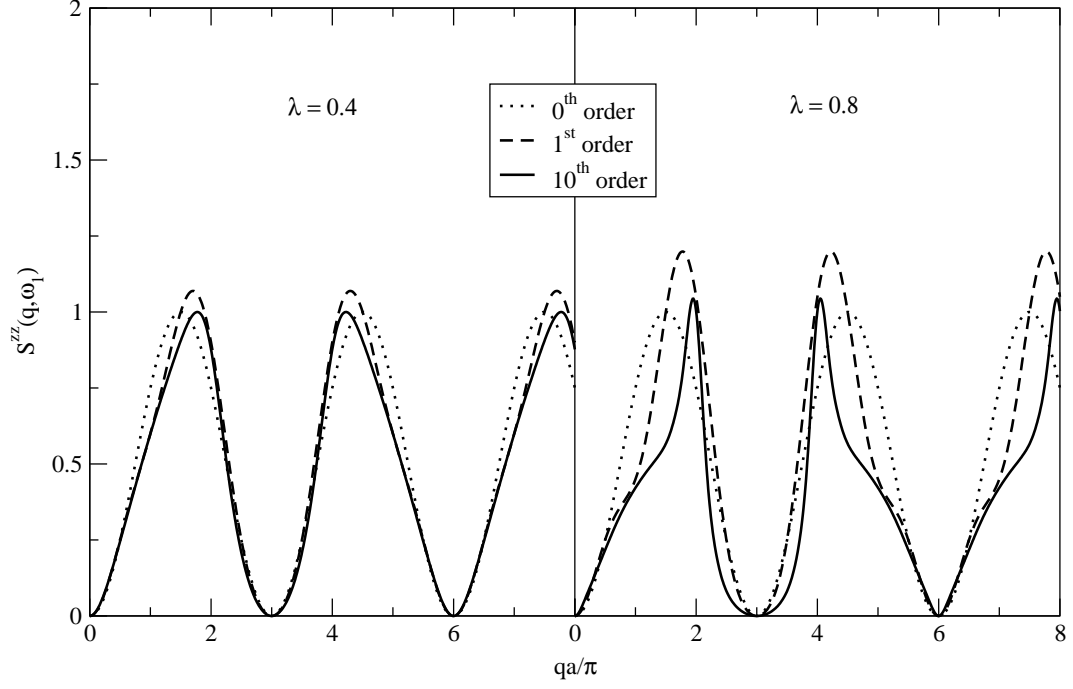


FIG. 3: Structure factor with $\lambda = 0.4, 0.8$ and ratio $d/a = 10/15$